# DENTAL RESTORATIVE COMPOSITE COMPOSITIONS AND FILLER THEREFOR

### BACKGROUND OF THE INVENTION

### 1. Field of the Invention

This invention relates to dental restorative composite compositions, and particularly, to the use of polyfluorocarbon resins or polychlorofluorocarbon resins, as a minor component of the finely divided, inorganic filler in dental restorative composite compositions.

#### 2. Discussion of the Prior Art

Dental restorative composites, generally in the form of highly filled blends of a liquid polymerizable organic resin matrix and finely divided, inorganic particulate 15 filler, have achieved wide commercial success and are used extensively in clinical dental practice. Basically, most of the dental restorative composites which have become commercially available, or which are described in the literature, are based upon the development of the  $\ ^{20}$ system first disclosed by Bowen in U.S. Pat. No. 3,066,112. In the direct filling system or restorative composite described in this patent, the liquid polymerizable organic resin matrix or binder is principally the reaction product of bisphenol and glycidyl methacry- 25 late, referred to as BIS-GMA, preferably combined with one or more other active monomers, referred to as reactive diluents, especially other dimethacrylates, for example, triethylene glycidyl methacrylate. The system also includes a catalyst or polymerization initiator, such 30 as, for example, benzoyl peroxide. Preferably, to allow the polymerization to take place in a reasonable period of time, a polymerization accelerator or activator, such as, for example, N,N-dimethyl-p-toluidine is also present in the composite. A particularly attractive catalyst- 35 /accelerator combination, which eliminates the colorforming amine, is cumene hydroperoxide-acetyl thiourea which is disclosed in U.S. Pat. No. 3,991,008. Other ingredients, such as stabilizers or UV-absorbers may also be present in association with the polymeriz- 40 able constituents to increase shelf life and otherwise prevent degradation of the properties of the restorative composite composition. Another ingredient usually used in dental restorative composite compositions is a coupling or fixing agent for enhancing the adhesion of 45 the inert inorganic filler particles with the binder matrix. Ethylenically unsaturated organosilane compounds are generally used for this purpose. Still further, restorative composite compositions may include various dyes or pigments to obtain various shades to conform to the 50 color of the tooth structure with which the restorative composite material is being used.

The composite restorative materials are generally provided for commercial use as multi-package systems, most typically a two package system, such as that described in U.S. Pat. No. 3,926,906 to Lee, et al. In these systems, the reactive monomers are generally provided in the form of a paste blended with the finely divided inert inorganic filler with the reactive diluent and/or catalyst and/or activator maintained separately from 60 each other and/or from the polymerization ingredients or reactive diluent.

The most commonly used inorganic filler materials are typically crystalline quartz or amorphous silica, although other materials, such as, for example, fused 65 silica, crystalline silica, glass beads, fused alumina and the like have also been disclosed. It has also been suggested to utilize fillers having a negative coefficient of

thermal expansion such as, for example, betaeucryptite, a lithium aluminum silicate. The use of fillers of low negative coefficients of thermal expansion is highly desirable in order to more closely match the resultant composite with the tooth structure in terms of thermal expansion. Still further, there have been several proposals for utilizing radio-opaque glasses as a component of the filler in dental restorative composite compositions. The use of radio-opaque additives enables the cured composite to be distinguished from the surrounding tooth structure during x-ray analysis commonly used in dental diagnostics.

While many advances have been made in the mechanical properties of dental restorative composite compositions, there does not seem to have been sufficient consideration given to the wear or abrasion resistance characteristics of the cured dental composites and there is still area for improvement in this particular property. The tooth-matching composite restoratives, as described above, are now widely used for interior restorations and repair of incisor fractures. In fact, their use for restorations where aesthetics is important has grown to an almost complete replacement of the previously used amalgams. However, composite restorations have been found in clinical studies to be unsuitable for occlusion surface restorations because of poor wear manifested by loss of anatomical form. Studies by Leinfelder, et al., J. Prosthet. Dent., 33, 407-416 (1975); Williams, et al., Int. Assoc. Dent. Res., Abst. No. 560, March 1972 and Phillips, et al., J. Prosthet. Dent., 30, 891-897 (1973) have shown that previously known dental composite resins have insufficient resistance to abrasive wear to be used in class I and II cavity preparations. Because of the poor clinical performance of composite resins compared to amalgams, the American Dental Association at present does not condone their use for occlusal surfaces. Thus, there is a great incentive to devise or produce a dental composite resin with resistance to wear abrasion that is equal to or superior to amalgams.

It has been suggested in U.S. Pat. No. 3,469,317 to Jarby that polyethylene, polymonochlorotrifluoroethylene or polytetrafluoroethylene, as well as several other synthetic polymers, in finely divided form, can be used directly as temporary or permanent tooth fillings or as a cavity lining for permanent fillings, and that such fillings exhibit a high resistance to abrasion and chewing pressure. Nevertheless, it has not been suggested that the finely divided polyhalocarbons, i.e. halogen-substituted polyethylenes, could be added, as only a minor amount of the total inorganic finely divided filler component of dental restorative composite compositions, and still impart such large improvement in abrasion resistance.

## **OBJECTS AND SUMMARY OF INVENTION**

It is accordingly an objective of the present invention to provide a dental restorative composite composition formed from a blend of liquid polymerizable resin binder matrix and a finely divided solid inert inorganic filler which has improved wear resistance, without loss of any other essential physical properties, such as compressive strength. It is also an object of the present invention to provide such dental restorative composite composition which can be used for occlusal surface restorations. These and other objects of the present invention will become more apparent from the following detailed description.